Relation between the wave front and the tip movement of spirals

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The relation between the movement of spiral waves and their tip for a simple spiral is experimentally investigated in a spatial open reactor using the Belousov-Zhabotinsky reaction. In the quasi-two dimensional regime, our results agree with the combustion model proposed by Lázár *et al.* [Chaos **5**, 443 (1995)] which states that the speed of spiral tip movement is proportional to the speed of spiral waves, $c_{tip} \propto c_{front}$, when expressed in the radius of the spiral tip trajectory R and the spiral wavelength λ, $R \propto \lambda$. The measurement shows that the slope of the linear R-λ curve is about 0.13, independent of the control parameter. This observation is also consistent with the combustion model.

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I. INTRODUCTION

Spiral patterns are some of the most intriguing spatiotemporal structures in macroscopic systems driven far from thermodynamic equilibrium. They are ubiquitous in different systems including reaction-diffusion media [1], aggregating slime mold cells [2], cardiac muscle tissue [3,4], and intracellular calcium release [5]. During the past 30 years, extensive theoretical and experimental studies have been conducted and fruitful results have been documented in the literature [6]. For example, we know that a spiral can be supported in excitable or oscillatory media, and is self-organized by a topological defect, that constructs the spiral core [1,6]; we have a deeper understanding of the nature of simple as well as meandering spiral waves [7–9], and we have identified several routes leading to spiral instabilities [10–14].

In a spiral-wave pattern, the pattern description is shown to naturally split between the tip region and far region; both parts are smoothly matched in an intermediate scale. A spiral tip is the wave source; a wave is sent out after the head of the tip moves one circle. Since the large scale motion of spiral waves is slaved to the spiral tip, the tip movement contains abundant information about the behavior of a spiral, and the spiral tip dynamics is possibly the key to understanding and controlling the behavior of spiral waves. Studies on the spiral tip movement, both in theory and in experiment, have generated rich information [7,8,15,16]. Several models have been proposed; some give good mathematical simulations of the meandering tip [8]; others furnish analysis on its movements near the core [15,16]. In this paper, we report on our experimental study of the relation between the radius of the spiral tip trajectory and the wavelength of the spiral waves in an excitable media using the Belousov-Zhabotinskii (BZ) reaction. After a description of the experimental setup, we present the result of the study. Our observation supports the combustion model proposed by Lázár *et al.* [16], which states that the speed of spiral tip movement is approximately equal to the speed of spiral waves. However, discrepancies exist, which call for further theoretical and experimental studies. A discussion is given at the end of the paper.

II. EXPERIMENTAL DESCRIPTION

We use a ferroin catalyzed BZ reaction to study the behavior of spiral waves. The experimental setup is the same as those used in early studies [17,18]. The reaction medium is a piece of porous glass disk, sandwiched between two reservoirs where the reactants of the BZ reaction are continuously refreshed and kept homogeneous by stirring. The chemicals are arranged in two reservoirs in such a way that one (A) is kept in the oxidized state of the reaction system and the other (B) in the reduced state of the reaction system. Pattern forming reactions take place in a thin layer inside the reaction medium when the reactants diffuse in the glass disk and meet together. Since there are multiple concentration gradients across the reaction medium, the observed pattern is quasi-three-dimensional, and instability in the gradient direction may arise. In this study, we focus on spiral patterns that are entrained in the gradient direction [19], so that the spirals can be considered as quasi-two-dimensional.

We choose the concentrations of sulfuric acid in reservoir A ($[H_2SO_4]_0^A$) and in reservoir B ($[H_2SO_4]_0^B$) as the control parameters, which are adjustable in a range of $0.4M$–$1.0M$ and $0M$–$1.0M$, respectively. Other conditions are kept fixed during the whole experiment: $[NaBrO_3]_0^{A,B}=0.2M$; $[CH_2(COOH)_2]_0^A=0M$; $[CH_2(COOH)_2]_0^B=0.6M$, $[KBr]_0^A=0M$, $[KBr]_0^B=60$ mM, $[\text{ferroin}]_0^A=1.0$ mM, and $[\text{ferroin}]_0^B=0M$. The resident time in each reservoir is 10^3 s; the ambient temperature is fixed at 25 °C.

The chemical patterns are monitored in transmitted light (halogen light filtrated to a wavelength less than 550 nm) with a charge coupled device (CCD) camera. The signal received by the CCD camera is sent to a computer where images are digitized and saved for further quantitative analysis. The initial condition of the experiment is that there is only one spiral tip residing in the middle of the reaction medium. This condition can be achieved in the following way: After suitable reactant solutions are pumped into reservoirs, a train of traveling waves automatically appears in the reaction medium. We use a beam of laser light (helium-neon laser, 3 mW, $\lambda=633$ nm) to break a chemical wave front and create

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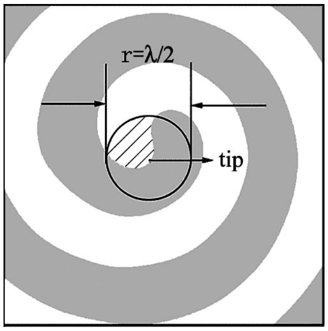

FIG. 1. Method to trace the trajectory of the spiral tip. The radius of the tool circle is specified as half of the wavelength, $r=\lambda/2$. The excited proportion (the shadowed part) reaches minimum at the tip.

a couple of defects, which develop into a pair of counter rotating spiral waves. Then we use laser light to lead one spiral tip to the edge of the reactor and delete it, and drive the other to the center of the reaction medium. Once a spiral is ready, it is studied by changing one reactant concentration stepwisely while fixing all other conditions. A series of pictures is taken after the pattern is relaxed into its asymptotic state, and no laser is applied for a sufficient amount of time (around an hour).

III. EXPERIMENTAL RESULTS

Because of its small size, the trajectory of a spiral tip is difficult to measure. In this experiment, we intend to study the movement of the spiral tip quantitatively. The aim of our effort is to investigate the relation between the wavelength of spiral waves and the radius of the circular trajectory of the spiral tip. From these data, we can deduct the relation between the speed of the spiral tip and that of the wave front. The wavelength is measured in the following way: In the region far from the spiral core, the Archimedean spiral is quite a good approximation of the configuration of spiral waves [20], so that we fit the curve of the spiral wave front with Archimedean's and look for the wavelength that best fits the observed data. Using this method, the relative error of the wavelength measurement is less than 1%. On the other hand, tracing down the trajectory of the spiral tips and calculating their radius of circular trajectory need more subtle procedures. Under our experimental conditions, the noise in the data has almost the same magnitude as the useful information. After comparing different methods, we found that the following procedures could give satisfactory results. First, we need to locate the spiral tip in each image. To obtain this information, we turn a digitized image into a black-white one with a specified threshold (usually the mean level of each image), then a tool circle, whose radius is a little less than the half-wavelength, is used to move along the black-white boundary of the spiral, with its center on the boundary (Fig. 1). At each step, the area of the white portion in the circle (the shadowed region in Fig. 1), corresponding to the area of the excited state in the system, is calculated and compared. The tip is defined as the location of the center where the value of the area reaches its minimum. This location approximately corresponds to the point of largest curvature of the wave front. Using this method the spiral tip of each image is marked, and the trajectory of the spiral tip is drawn. Figure 2 gives examples of simple spirals with the trajectory of their tips. One observes that the trajectory follows a circle. We then fit the trajectory of the spiral tip with a ring, and get the data of its radius.

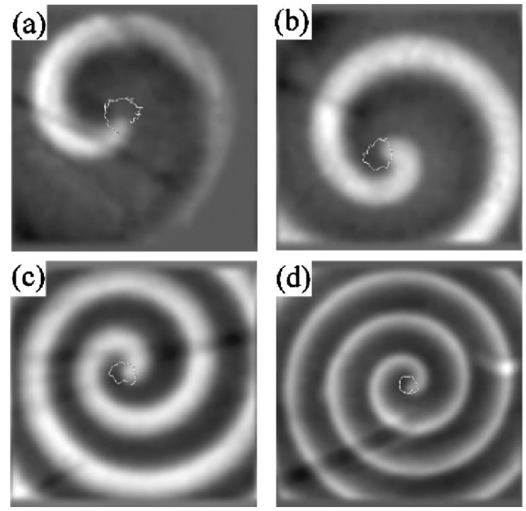

FIG. 2. Examples of spiral patterns and spiral tip trajectories with different control parameters. The values of control parameters $[H_2SO_4]^A$, and $[H_2SO_4]^B$ and the measurements of λ and R are in (a) 0.4M, 0.1M, 0.974 mm, and 0.058 mm; in (b) 0.7M, 0.067M, 0.654 mm, and 0.05 mm; in (c) 0.8M, 0.6M, 0.415 mm, and 0.042 mm; and in (d) 1.0M, 1.0M, 0.295 mm, and 0.033 mm.

Once we have traced the trajectories of the spiral tips, we distinguish the dynamical behavior of the system according to the spiral tip movements. Figure 3 shows the phase diagram. We found that when the concentration of sulfuric acid in the reduced side ($[H_2SO_4]^B$) is about 0.25M larger than that in the oxidized side ($[H_2SO_4]^A$), the spiral patterns can-

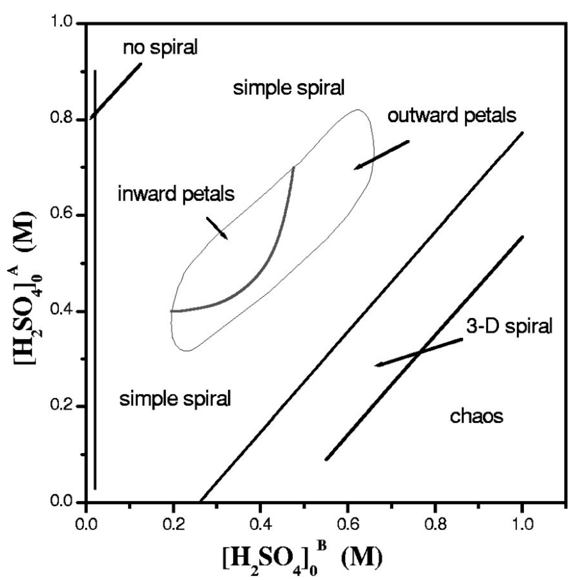

FIG. 3. Phase diagram of the system in the subspace of control parameters ($[H_2SO_4]^A$ and $[H_2SO_4]^B$).

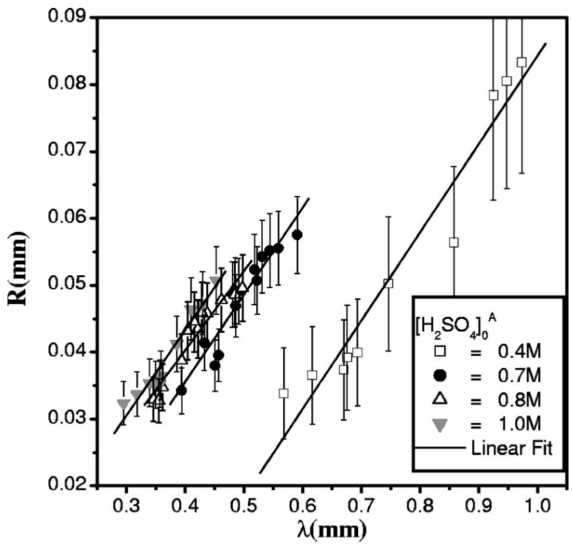

FIG. 4. Relation between the radius of spiral tip trajectory R and the wavelength λ. R is expressed as a function of λ. The error bar is 10% for $[H_2SO_4]^A = 0.7M$, $0.8M$, and $1.0M$ and 20% for $[H_2SO_4]^A = 0.4M$.

not be described as quasi-two-dimensional. A clear signature of three-dimensional (3D) structure is observed. This 3D effect can induce spiral instability when the system crosses the turbulence line in Fig. 3. A detailed description of this type of transition to spatiotemporal chaos will be reported elsewhere. The states in the left part of the phase diagram in Fig. 3 can be described as quasi-two-dimensional spirals that include simple spiral and meandering spirals [8,9]. Different from the previous study [9], which uses $[CH_2(COOH)_2]^B$ and $[H_2SO_4]^B$ as the control parameters, in this study the codimension-2 points separating the outward-petal and inward-petal meandering spirals are not located at the climax of the meandering boundary.

The relation between the wavelength (λ) and the radius of the spiral tip trajectory (R) is studied in the simple spiral regime in Fig. 3. We measure the values of λ and R using the method described earlier, and find that, for a given value of $[H_2SO_4]^A$, the wavelength of spiral waves is always nearby proportional to the radius of spiral tip trajectory as the control parameter $[H_2SO_4]^B$ is varied, as shown in Fig. 4. There is no apparent difference in the spiral tip behavior between left and right sides of meandering region. We deduce a linear relation between λ and R, although some disharmony exists. The slope and the interception of the lines is (0.13, -0.05), (0.13, -0.02), (0.12, -0.01), and (0.13, -0.01) for $[H_2SO_4]^A = 0.4M$, $0.7M$, $0.8M$, and $1.0M$, respectively. The error bar for $[H_2SO_4]^A = 0.7M$, $0.8M$, and $1.0M$ is about 10%, while for $[H_2SO_4]^A = 0.4M$, it is larger (about 20%). Although the unshakable error is often in the same level as the useful information in the measurement of spiral tip position, when we fit all the locations with a ring, which is equivalent to doing an averaging, the error bar comes down and the result can be accepted as a quantitative one.

IV. DISCUSSION

Lázár *et al.* have described a model of spiral movement using an analogy of the combustion of grass [16]. Adopting this model, we deduce that for the first-order approximation the following relation holds for a state of simple spirals:

$$c_{tip} \approx c_{front}, \tag{1}$$

where c_{tip} and c_{front} denote the tangential traveling velocity of the spiral tip and the speed of the wave fronts in the normal direction, respectively. When expressed in terms of the radius of the spiral tip trajectory R and the spiral wavelength λ, the relation becomes

$$R \propto \lambda_{front}, \tag{2}$$

with a slope of $1/2\pi = 0.159$.

Our observation supports the prediction of the linear relationship between R and λ, and in a first-order approximation, the value of the slopes also agrees with the theoretical value ($1/2\pi \approx 0.159$). Furthermore, according to the theoretical prediction, the slope of the linear relation between λ and R should be independent of control parameters; our experimental results confirm this prediction.

However, an apparent discrepancy exists. As shown in Fig. 4, the interception value for $[H_2SO_4]^A = 0.4M$ is -0.05, clearly away from 0. Extrapolating the relation to $R = 0$, one expects that even when the spiral tip does not move, spiral waves with a certain wavelength exist. This result is inconsistent with the combustion model. At present, we do not have a sure theoretical explanation of this experimental result. We speculate that the combustion model is only a first-order approximation. Other factors must be considered in order to get a full picture of the spiral dynamics.

From the experimental point of view, it is difficult to deduce an accurate quantitative relation between the radius of the spiral tip trajectory and the wavelength of spiral waves. The reasons lie in three aspects. First, there is only a very narrow range of control parameters that allows simple spiral waves to exist, restricted by the tendency of spiral tip meandering and 3D spiral structures (see Fig. 3). In this narrow range, the variation of the core size is very small, which leads to a large error bar. Second, large relative errors exist in the measurement of the position of a spiral tip. The edge of the excited region in a spiral is often not sharp enough for us to locate the spiral tip exactly, thus the standard error of the measurement often almost overshadows the circle size itself. Various methods have been tried to overcome this difficulty; only limited improvement has been achieved. Third, the definition of spiral tip is artificial. Different definitions of spiral tip exist [7,8,21]. This does not affect *qualitative* conclusions, i.e., $R \propto \lambda_{front}$. However, since we are interested in the *quantitative* relation between the trajectory radius and the wavelength, the exact definition of the tip must be provided, because different definitions give quite different results. If we consider the definition that defines the tip as the point where the wave front meets the wave back [21] (this definition is being used widely in the literature), the position of a spiral tip should be the point of zero curvature on the boundary of the excited state. In this case, our study shows that the measured values of R will be about 20–25 % higher, which leads to the value of slope of about 0.16. This value is very close to the theoretical prediction of $1/2\pi = 0.159$. However, in the experiment we did not use this definition because getting the value of curvature involves calculating the second derivative of the wave fronts. The noise of original data will be largely amplified in this operation, introducing a huge error bar.

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